



Pyrethroid insecticides in urban catch basins: A potential secondary contamination source for urban aquatic systems[☆]

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ABSTRACT

Pesticide contamination is a threat to many aquatic habitats, and runoff from residential homes is a major contributor of these chemicals in urban surface streams and estuaries. Improved understanding of their fate and transport can help identify areas of concern for monitoring and management. In many urban areas, runoff water congregates in numerous underground catch basins before draining into the open environment; however, at present essentially no information is available on pesticide presence in these systems. In this study, we collected water samples from a large number of underground urban catch basins in different regions of California during the active pest management season to determine the occurrence and profile of the widely used pyrethroid insecticides. Detectable levels of pyrethroids were found in 98% of the samples, and the detection frequency of individual pyrethroids ranged from no detection for fenprothrin to 97% for bifenthrin. In the aqueous phase, total pyrethroid concentrations ranged from 3 to 726 ng/L, with a median value of 32 ng/L. Pyrethroids were found to be enriched on suspended solids, with total concentrations ranging from 42 to 93,600 ng/g and a median value of 2,350 ng/g. In approximately 89% of the samples, whole water concentrations of bifenthrin were predicted to have toxic units >1 for sensitive aquatic invertebrates. The high detection frequency of bifenthrin and overall pyrethroid concentrations, especially for particle-bound residues, suggest that underground urban catch basins constitute an important secondary source for extended and widespread contamination of downstream surface waters by pesticides such as pyrethroids in urban regions.

1. Introduction

Pesticides are man-made chemicals with biological activity. Off-site transport commonly occurs due to widespread use and various environmental processes, leading to contamination in sensitive ecosystems. Compared to agricultural fields, relatively little is known about the urban fate and movement of pesticides, although a wide range of pesticides are routinely used for pest management of structures and

landscapes. Pyrethroids, one of the most applied types of insecticides in urban settings, are used broadly for managing nuisance pests and vectors such as ants, spiders, cockroaches, and mosquitoes. In California alone, reported use of seven pyrethroids for structural pest control and landscape maintenance was over 70,000 kg (as active ingredient) in 2017 (Budd et al., 2020; "California pesticide information portal application," 2022). This was likely a significant underestimate, as products containing pyrethroids are readily available to homeowners through retail

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stores and online sites.

Urban areas differ characteristically from agricultural fields in that there is a prevalence of impervious surfaces on which pesticide residues may accumulate (Jiang et al., 2016; Richards et al., 2016). Surface runoff water after rainfall or irrigation events moves swiftly over impervious surfaces and may mobilize the deposited residues. For example, a study of simulated runoff from concrete slabs showed detectable levels of pyrethroids in runoff water 112 d after the treatment (Jiang et al., 2010). Studies over recent years has implicated surface runoff for contamination of pyrethroids and other pesticides in urban water bodies (Weston et al., 2009). Many studies have shown occurrence of pyrethroids in creeks and rivers in urban watersheds, with concentrations in the water column or bed sediment frequently exceeding toxicity benchmarks for aquatic invertebrates (Ensminger et al., 2013; Weston et al., 2009; Weston and Lydy, 2012). More recently, Budd et al. (2020) reported that 78% of surface water samples from urban watersheds across 8 California counties contained one or more pyrethroids, with bifenthrin being the predominant compound. Pyrethroid contamination has also been reported for urban sites in other states, including Texas and Illinois, and other countries, including China, Vietnam, and Australia (Allinson et al., 2015; Ding et al., 2010; Duong et al., 2014; Hintzen et al., 2009; Kuivila et al., 2012; Tang et al., 2018).

Pyrethroid exposure poses a risk to many non-target species, such as crustaceans, aquatic insects, amphibians, and fish, and their habitats may suffer a loss of biodiversity and ecosystem functions (Haya, 1989; Siegfried, 1993; Vanzetto et al., 2019). For example, a study of urban streams and associated outfalls in Oregon, U.S.A., showed the presence of over 30 pesticides, of which bifenthrin was found at the highest levels in the sediment and the contamination was associated with impairment of invertebrate communities (Carpenter et al., 2016). The strong hydrophobic nature of pyrethroids results in their accumulation in sediment, where pyrethroids may exhibit a prolonged persistence due to slow degradation, especially under anaerobic conditions (Gan et al., 2005; Laskowski, 2002; Meyer et al., 2013).

Before entering surface waters, urban runoff drainage generally passes through an underground storm drain system (USDS). These systems, designed to limit flooding and stormwater pollution, consist of underground catch basins, pipes, and channels. The USDS can be extremely extensive in highly urbanized areas. For example, the USDS of

the City of Los Angeles, CA, comprises of over 38,000 catch basins, 2,414 km of pipes, and 161 km of open channels, with estimated annual dry weather and wet weather runoffs of 189,250 m³ and 38 million m³, respectively (Sadeghi et al., 2017). On a broader scale, Los Angeles County has over 70,000 catch basins, 7,400 km of sewer pipes, and 1,340 km of surface channels (Kwan et al., 2010; Porse, 2018). Catch basins, usually located curbside, accept runoff from the paved surface. Most catch basins contain a sump that can temporarily hold water and debris and a connecting pipe allowing water to enter the USDS (Fig. 1). Catch basins are usually concrete-lined and rectangular in shape with a depth of up to several meters (Anderson et al., 2011; Harbison et al., 2018).

There have been a few studies on stormwater structures concerning nutrients, such as nitrate and phosphorus, and conventional contaminants, such as PAHs and metals (Azah et al., 2017; Eriksson et al., 2007; Lundy et al., 2012). Modeling studies have also considered sediment capture dynamics and contaminant removal (Alam et al., 2017, 2018b; Jang et al., 2010; Morgan et al., 2005; Yang et al., 2018). Despite the magnitude of urban USDS systems and their significance in transporting urban runoff, there is a dearth of information on contaminant (including pesticides) fate and transport through these underground systems before discharge into the open environment.

The current study evaluated the occurrence of the commonly used pyrethroid insecticides in urban catch basins to improve understanding of the role of USDSs in the overall contamination of urban watersheds. Their large number and broad distribution, along with their unique environmental settings (e.g., underground, dark, lower temperature) imply that catch basins may significantly affect contaminant movement and distribution by first intercepting and then serving a secondary source for contaminant emission. Insights into pyrethroid environmental behavior may also be useful for predicting that of other strongly hydrophobic compounds in the urban environment.

2. Materials and methods

2.1. Chemicals and materials

Analytical standards of pyrethroids were obtained from various pesticide manufacturers. Tefluthrin (94.0% purity) and lambda-

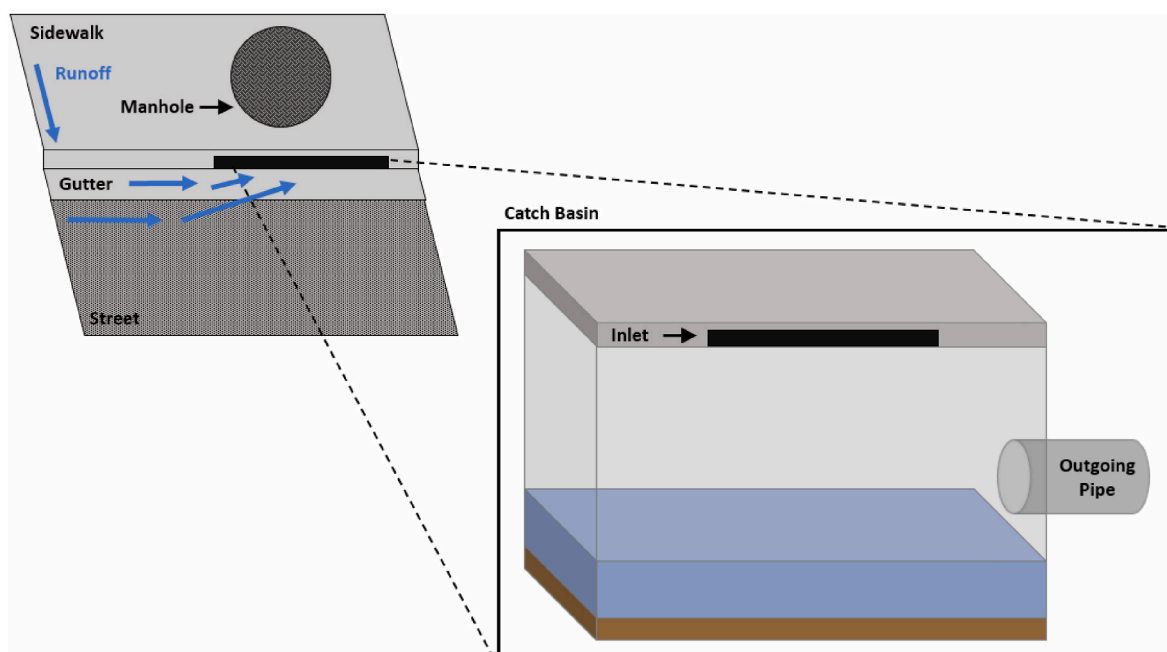


Fig. 1. Diagram of runoff flow into a catch basin. Runoff enters through a roadside inlet and may accumulate in the basin's sump before flowing out through a pipe.

cyhalothrin (89.0%) were obtained from Syngenta Crop Protection (Greensboro, NC). Bifenthrin (98.4%), permethrin (98.5%), and cypermethrin (98.5%) were obtained from FMC Corporation (Philadelphia, PA). Fenpropathrin (99.7%) and esfenvalerate (99.9%) were obtained from Valent (Dublin, CA). Cyfluthrin (86.4%) and deltamethrin (99.4%) were obtained from Bayer Crop Science (Research Triangle Park, NC). Physicochemical properties for select pyrethroids are listed in [Table S2](#). Deuterated bifenthrin- d_5 used to determine surrogate recovery was purchased from MilliporeSigma (St. Louis, MO).

Dippers and glass bottles for water sample collection were purchased from John W. Hock Company and J.G. Finneran Associates, respectively (Gainesville, FL; Vineland, NJ). pH buffer solutions for calibration were purchased from Thermo Fisher Scientific (Waltham, MA). Total dissolved solid standards were purchased from Oakton Instruments (Vernon Hills, IL). Glass fiber filters were purchased from Advantec MFS (Dublin, CA). Sodium sulfate, sodium chloride, and GC/MS grade solvents (dichloromethane, acetone, and hexane) were purchased from Thermo Fisher Scientific (Waltham, MA). Florisil was purchased from Spectrum Chemical (New Brunswick, NJ). Before use, Florisil was activated via baking at 130 °C for 4 h, and sodium sulfate was dehydrated via baking at 400 °C for 4 h. All glassware used for sample processing was baked at 400 °C for 4 h beforehand to prevent cross contamination.

2.2. Site selection and sampling

Much of California, including the regions where catch basins were sampled, falls under a Mediterranean climate, characterized by dry

summers and infrequent storm events during the winter (Polade et al., 2017). The majority of the state's rainfall occurs from November through March, with very little rainfall observed May through September (Fierro, 2014). Thus, it is assumed that water in urban catch basins over the summer is predominantly, if not exclusively, from irrigation runoff.

The catch basins were selected from locations in 5 northern California counties (Sacramento, Yolo, Alameda, Madera, and Tulare) and 3 southern California counties (Los Angeles, Orange, and San Bernardino), therefore representing a large portion of the urbanized areas in California ([Fig. 2](#)). Site selection and sampling was performed in collaboration with regional agencies, including Sacramento-Yolo Mosquito and Vector Control District (MVCD), Alameda County Mosquito Abatement District, Madera County MVCD, Delta MVCD, Greater Los Angeles County Vector Control District, Orange County MVCD, and West Valley MVCD. Vector control agencies in California regularly access catch basins for surveillance and management of mosquitoes. Sampling sites consisted of 8–13 urban catch basins in each sampling region (see [Table S1](#) for site details). When possible, additional catch basins were sampled to make up for any sites that were dry at the time of sample collection.

Each catch basin with water was sampled once a month from July to September 2020. Telescoping dippers with coated stainless-steel cups were used to transfer approximately 500 mL of water into 16 oz amber glass bottles. Care was taken to collect from near the surface of the water to avoid disturbing bottom sediment or debris. Samples were immediately chilled and transported to the University of California, Riverside.



Fig. 2. Counties in California where catch basins were sampled. Red dots represent areas where catch basins were sampled. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Upon receipt at the laboratory, samples were stored at 4 °C in the dark prior to processing and analysis.

2.3. Sample preparation

A Fisherbrand AE150 pH meter was used to measure sample pH before filtration (Waltham, MA). To separate suspended solids, each whole water sample was vacuum filtered through a 0.4 µm pore size glass fiber membrane (Advantec MFS, Dublin, CA). After filtration, a 20 mL aliquot of water was taken to measure total dissolved solids (TDS) and dissolved organic carbon (DOC) with a YSI EcoSense EC30A conductivity pen (Yellow Springs, OH) and a Shimadzu TOC-V analyzer, respectively (Columbia, MD). Filters with suspended solids were covered with aluminum foil and stored in a refrigerator at 4 °C until extraction.

After weighing, water samples were extracted using liquid-liquid extraction. Briefly, in a 2 L glass separatory funnel, 30 g of sodium chloride was added to the 500 mL water sample and then shaken until dissolved. Bifenthrin-d₅ (100 ng in 100 µL acetone) was added to each sample as a recovery surrogate. For extraction, 60 mL dichloromethane was added to each sample, and then the separatory funnel was mixed vigorously by hand for 3 min and left to rest for 1 h. After phase separation, the solvent extract was drained into a round-bottom glass flask through a glass funnel packed with anhydrous sodium sulfate (30 g) to remove residual water. This extraction was repeated for a total of three consecutive times, and the extracts were combined for each sample. The combined extract was then condensed on a Buchi Rotovapor (New Castle, DE) at 40 °C and under vacuum to approximately 5 mL. The sample container was rinsed with 9:1 hexane:acetone (v/v) and the extract was transferred to a glass vial. The final sample was evaporated to near dryness at 40 °C under a gentle stream of nitrogen and then reconstituted in 1.0 mL hexane for analysis via gas chromatography/mass spectrometry (GC/MS).

To extract pyrethroids from the suspended solids retained on the filter paper, the sample filter membranes were first dried in an oven at 105 °C for 24 h. Total suspended solids (TSS) were measured gravimetrically by weighing the dried filters. Afterwards, samples were placed in a 40 mL glass vial and spiked with bifenthrin-d₅ (100 ng in 100 µL acetone). For extraction, 10 mL of 1:1 hexane:acetone (v/v) was added to each sample, and then the samples were sonicated for 30 min in a sonication water bath. For cleanup, the extract was loaded onto a 1.0 g Florisil cartridge preconditioned with 6 mL hexane, and the cartridge was eluted with 10 mL of 9:1 hexane:acetone (v/v). The cleaned extract was evaporated to near dryness at 40 °C under a gentle stream of nitrogen and then reconstituted in 1.0 mL hexane for GC/MS analysis.

2.4. Chemical analysis and quality control

Sample extracts were analyzed for tefluthrin, bifenthrin, fenprothrin, lambda-cyhalothrin, permethrin, cyfluthrin, cypermethrin, esfenvalerate, and deltamethrin on an Agilent 6890 N/5973B GC/MSD operated in electron ionization mode with a 30 m × 0.25 mm × 0.25 µm DB-5MS column. The GC oven temperature program was as follows: initial temperature 50 °C held for 1 min; heated to 220 °C at 30 °C/min; heated to 300 °C at 10 °C/min and held for 2 min; and post run hold at 310 °C for 5 min. Injection volume was 1.0 µL and carrier gas (helium) flow rate was set to 1.0 mL/min. Temperatures for the injector, transfer line, MS source, and MS quadrupole were 250 °C, 280 °C, 230 °C, and 150 °C, respectively. A solvent delay of 6 min was set for protection of the MS filament. Quantitation was performed with an 8-point calibration curve of concentrations from 1 to 500 µg/L.

For quality control and assurance, a calibration standard was run for every 10 samples to check for potential instrumental drift. Relative standard deviations were found to range from 6% to 14%. Bifenthrin-d₅ was added to each sample as a surrogate to measure recovery. The average recovery was 99 ± 18% and ranged from 72% to 127%. Method

extraction efficiency was determined from the analysis of triplicate samples of water or sediment spiked with the target analytes. Method blanks simultaneously extracted with samples showed negligible matrix effects. Reagent blanks, analyzed after every 10 samples, showed no presence of the target analytes.

Method detection limit were determined following EPA method 40 CFR, Part 136, Appendix B. Reporting limits were defined as 3 times the method detection limits. Detailed analyte MS parameters, reporting limits, and method extraction efficiencies are given in Table S3. Detection frequencies were calculated using the number of samples in which residues were found above the reporting limit in either the aqueous phase or suspended solids. Non-detects and concentrations below the reporting limit were considered 0 when calculating values from all sample data. All concentrations in suspended solids were reported on a dry weight basis. The partitioning coefficient K_d was calculated using the formula C_s/C_w , where C_s and C_w are the concentrations in the solid and aqueous phases, respectively. Using quantified pyrethroid concentrations and other environmental data, linear regressions were performed to determine whether variables, including pyrethroid concentrations, pyrethroid partitioning across media, TSS, and DOC, were correlated with one another.

3. Results and discussion

3.1. Detection frequencies and concentrations

A total of 79 urban catch basins in California were sampled from July to September 2020, months during which runoff would have been driven by irrigation only. Of these basins, 58 were sampled for all 3 months, showing consistent retention of water. The results showed that 98% of catch basin water samples contained at least one pyrethroid compound at a concentration above the reporting limit (Table S4). Bifenthrin consistently exhibited the highest frequency of detection, being detected in 97% of the samples. In previous studies on dust around residential homes, urban runoff, and urban surface waters receiving urban drainage in California, bifenthrin was often found at the highest frequency among urban-use pesticides (Budd et al., 2020; Carpenter et al., 2016; Ensminger et al., 2013; Holmes et al., 2008; Jiang et al., 2016; Richards et al., 2016). This may be attributed to the widespread use of bifenthrin-containing products in urban environments and also its relatively long persistence in the environment (Gan et al., 2005; Meyer et al., 2013). According to the California Department of Pesticide Regulation's (CDPR's) pesticide use databases, bifenthrin was the most applied pyrethroid for non-agricultural landscape maintenance and structural pest control in many regions in the state ("California pesticide information portal application," 2022).

Lower detection frequencies were observed for other pyrethroids (Table S4). The second most frequently detected pyrethroid was lambda-cyhalothrin, which was found in about 30% of the water samples. Cyfluthrin, permethrin, esfenvalerate, cypermethrin, and deltamethrin had overall frequencies of detection of 16%, 8%, 7%, 4%, and 1%, respectively. In previous studies, permethrin and cyfluthrin were associated with toxicity in several impaired waters, though not as frequently as bifenthrin (Budd et al., 2020; Holmes et al., 2008; Jiang et al., 2016; Richards et al., 2016; Weston et al., 2009). Fenprothrin and tefluthrin were not detected in any of the samples in this study. Frequencies of detection between the aqueous and suspended solid phases were nearly identical, although there were a few exceptions. For example, bifenthrin was detected in the filtered water (i.e., aqueous phase) in the Sacramento-Yolo region at 86%, but at 97% in the suspended solids (n = 36). This small discrepancy was likely due to the different reporting limits for the aqueous and solid phase samples.

Pyrethroid concentrations in catch basin water (C_w) and suspended solids (C_s) are summarized in Tables 1 and 2. Except for bifenthrin, median concentrations for pyrethroids were below reporting limits in both aqueous phase and suspended solids. Total concentrations of the

Table 1
Summary of pyrethroid concentrations in aqueous phase of catch basin water (n = 205).

Pyrethroid	Detection frequency (%)	Median (ng/L)	Max. (ng/L)	75th percentile (ng/L)
Tefluthrin	0	<RL	<RL	<RL
Bifenthrin	90	20	324	31
Fenpropathrin	0	<RL	<RL	<RL
Lambda-cyhalothrin	25	<RL	626	<RL
Permethrin ^a	1	<RL	20	<RL
Cyfluthrin ^a	12	<RL	689	<RL
Cypermethrin ^a	3	<RL	253	<RL
Esfenvalerate ^a	4	<RL	127	<RL
Deltamethrin	1	<RL	85	<RL
All pyrethroids	92	32	726	87

RL = reporting limit.

^a Quantified as the sum of isomers.

Table 2
Summary of pyrethroid concentrations in suspended solids from catch basin water (n = 205). Concentrations are expressed on a dry weight basis.

Pyrethroid	Detection frequency (%)	Median (ng/g)	Max. (ng/g)	75th percentile (ng/g)
Tefluthrin	0	<RL	<RL	<RL
Bifenthrin	97	1440	75400	3190
Fenpropathrin	0	<RL	<RL	<RL
Lambda-cyhalothrin	30	<RL	18200	<RL
Permethrin ^a	8	<RL	76100	<RL
Cyfluthrin ^a	16	<RL	39800	<RL
Cypermethrin ^a	4	<RL	5690	<RL
Esfenvalerate	7	<RL	8430	<RL
Deltamethrin	1	<RL	7230	<RL
All pyrethroids	98	2350	93600	5910

RL = reporting limit.

^a Quantified as the sum of isomers.

target pyrethroids in water ranged from 3 to 726 ng/L, with a median of 87 ng/L. For suspended solids, they ranged from 42 to 93,600 ng/g, with a median of 2,350 ng/L. Bifenthrin was detected at up to 324 ng/L in water and 75,400 ng/g in suspended solids. The maximum concentration of bifenthrin found on the suspended solids was significantly higher than those seen in previous studies. For example, in Weston et al., the total concentrations of pyrethroids in suspended solids ranged from 1,150 to 3,390 ng/g in water samples collected at stormwater outfalls in northern California (Weston et al., 2009). It must be noted that the high concentrations observed in the current study were only from a few samples, and the same catch basins showing high concentrations in a single sampling event did not show elevated levels at the other sampling time points. This may indicate a recent pesticide application in the vicinity of the specific catch basins, causing transient high pesticide loadings.

While bifenthrin was the most frequently detected pyrethroid in each region or month, other pyrethroids often made up a greater proportion of a region’s total pesticide concentrations (Fig. 3). For example, in the August samples collected from Los Angeles County and the September samples from the West Valley region, lambda-cyhalothrin accounted for 88% of concentrations and permethrin accounted for 91% of concentrations, respectively. When considering the entire sampling period, the only region where the bifenthrin concentration was dominant was Madera County in northern/central California. Overall, bifenthrin represented 44% of average total pyrethroid concentrations, which was smaller compared to that (72%) observed in Budd et al. (2020) for surface waters across California.

3.2. Patterns and influencing factors

Median total pyrethroid concentrations among the different sampling regions ranged from 20 ng/L to 64 ng/L in the aqueous phase and 946 ng/g to 3,890 ng/g in the solid phase (Fig. 4). Though differences can be observed in concentrations among different regions, these trends cannot be extrapolated broadly given the limited number of sampling locations in each region. In California, reported use of pyrethroid insecticides differs widely by county, as counties have different populations, population densities, climate conditions, and land use patterns.

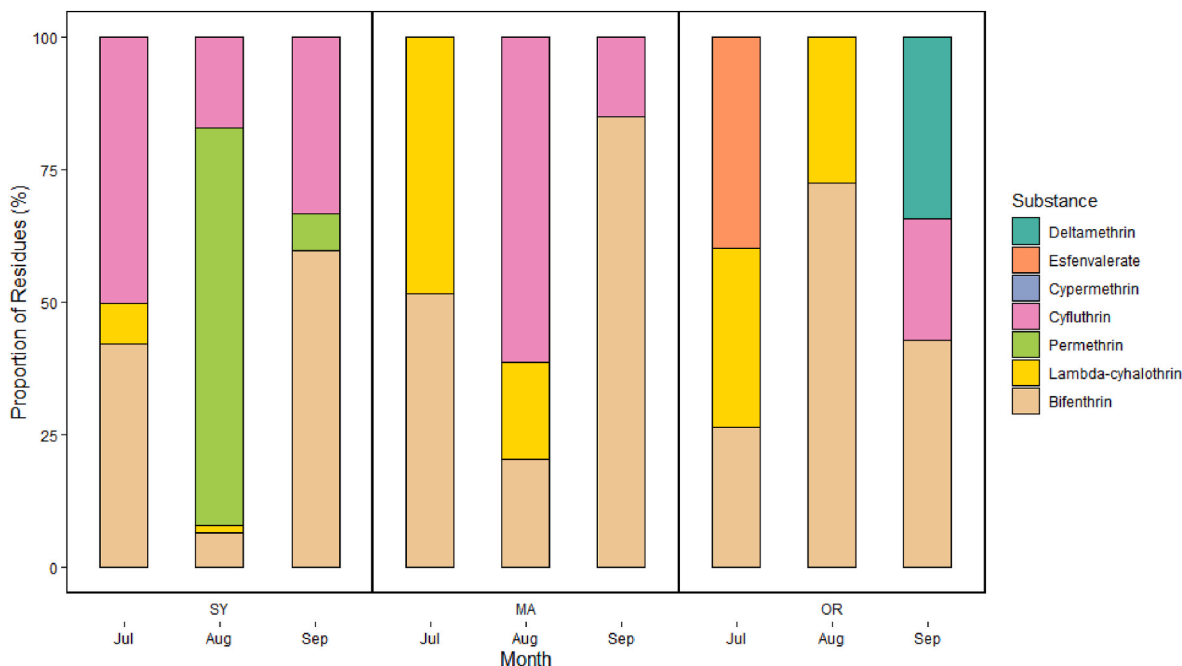


Fig. 3. Fractions of individual pyrethroids in whole water samples for three sampling sites (Sacramento/Yolo Counties = SY; Madera County = MA; Orange County = OR). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

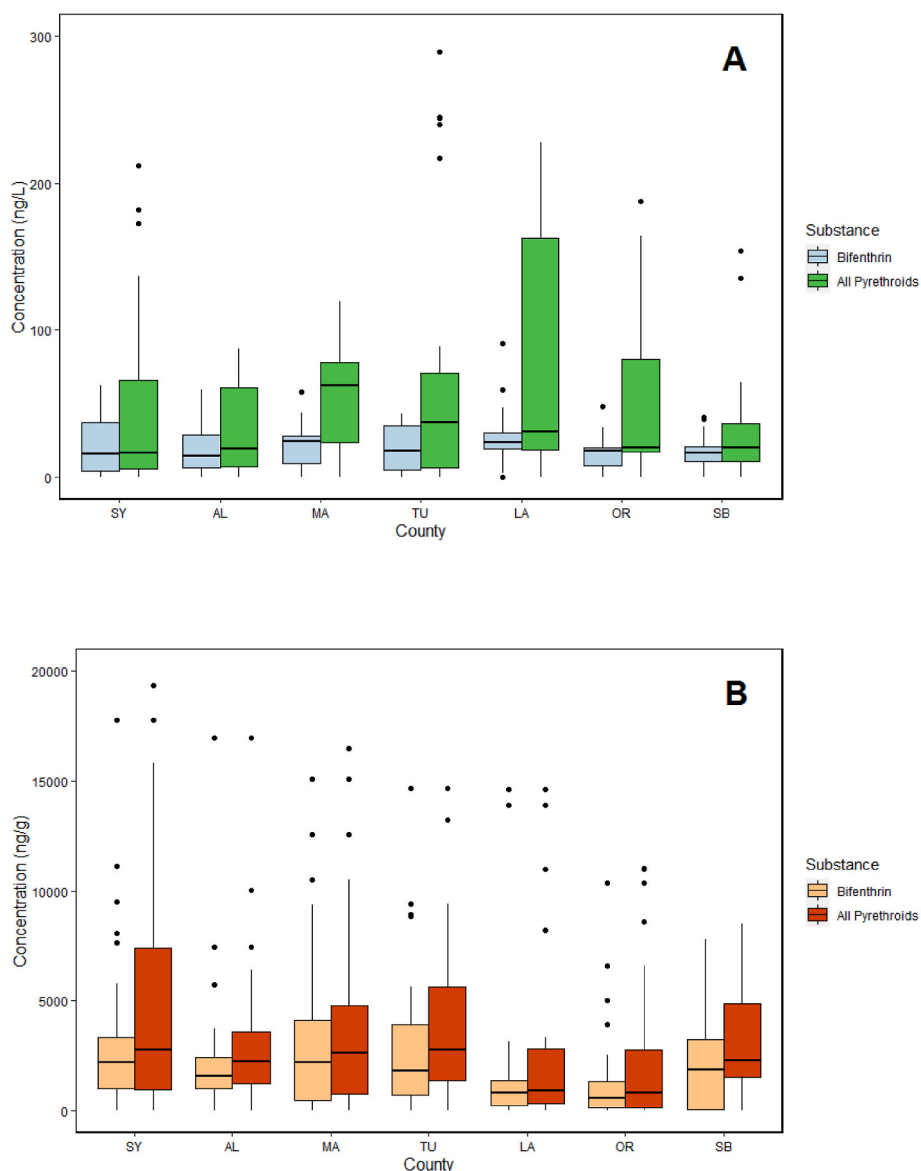


Fig. 4. Bifenthrin and total pyrethroid concentrations in A) aqueous phase and B) suspended solids; grouped by sampling region. Sacramento/Yolo Counties = SY; Alameda County = AL; Madera County = MA; Tulare County = TU; Los Angeles County = LA; , Orange County = OR; and San Bernardino County = SB. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Previous studies have reported higher detection frequencies in Southern California (e.g., Orange County) than in Northern California for pyrethroids and other pesticides (e.g., fipronil), but this trend was not observed in the catch basins surveyed in this study (Ensminger et al., 2013; Gan et al., 2012). (Budd et al., 2020) noted that flood control channels in Southern California are more commonly lined, increasing water-associated contaminant transport; this may partially explain why downstream detections and concentrations are more disparate between regions as compared to those from roadside catch basins. Other regional differences, such as local pest types and pressures (which may be influenced in turn by factors like climate and vegetation), management practices, and regulations, can significantly affect pesticide use and thus their offsite transport.

Median concentrations in both water and suspended solids decreased from July to September (Fig. 5). The median total pyrethroid concentrations in July, August, and September for water were 85 ng/L, 24 ng/L, and 6 ng/L, respectively, and those for suspended solids were 2,780 ng/g, 2,480 ng/g, and 1,260 ng/g, respectively. Much of this trend was driven by decreases in bifenthrin concentrations, with median

concentrations in the aqueous phase decreasing from 28 ng/L in July to 5 ng/L in September and concentrations in suspended solids from 2,180 ng/g in July to 750 ng/g in September.

Pesticide use data from CDPR’s Pesticide Information Portal were taken into consideration to understand the trends of pyrethroids seen in the catch basins (“California pesticide information portal application,” 2022). The trend in monthly reported urban use of pyrethroid insecticides varies among counties. For example, Sacramento County use of the pyrethroids in this study was highest in January, November, and December, Yolo County use was highest in the summer months (with a peak in July), and San Bernardino County use remained relatively consistent throughout the year. In a few counties, use in one month was much higher than in others; for example, in Tulare County, use in April was 423 kg, which was almost three times the monthly average of 146 kg for the year. In addition, winter rain events likely wash off a large amount of pyrethroid residues (relative to irrigation-induced runoff) into the catch basins, and the concentrations decrease with time during the dry season, which further complicates the response of pesticide occurrence in the catch basins to pesticide use. The suitability of

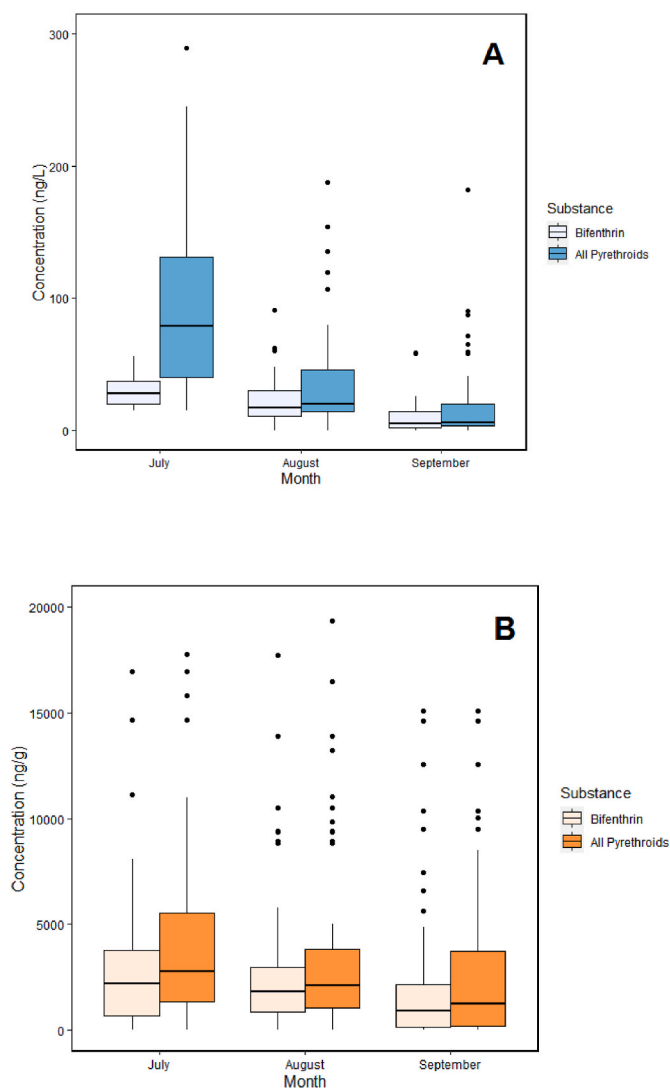


Fig. 5. Bifenthrin and total pyrethroid concentrations in A) aqueous phase and B) suspended solids (grouped by sampling month).

predicting and understanding trends in the environmental presence of pyrethroid residues from pesticide use data is further impacted by the difference in persistence of individual compounds. Pyrethroids are known to be extremely hydrophobic, and once entering a catch basin, are quickly partitioned into the bottom sediment. The redistribution from the sediment phase to water may serve as a continuous secondary source discharging pyrethroid residues into downstream surface water systems on a prolonged time scale (Gan et al., 2005; Meyer et al., 2013).

Water chemistry variables such as TSS and DOC are known to affect the behavior of stormwater contaminants, especially strongly hydrophobic compounds like the synthetic pyrethroids. Mean TSS in water samples was 39 ± 80 mg/L (Table 3) and the median TSS was 11 mg/L, showing wide variance in TSS levels among different catch basins as well as the same basins at different sampling times. This mean value is similar to those in urban runoff from outfalls in Aliso Viejo, CA, during the dry season (41 mg/L) and from a stormwater channel flowing into the San Francisco Bay during low flow conditions (68 mg/L) (Gilbreath and McKee, 2015; Pitton et al., 2016). However, TSS differs significantly across previous literature and may be influenced by a combination of seasonal and geographic variables. Mean TSS from some outfalls during the dry season was as low as 0.55 mg/L, while the Environmental Protection Agency's National Urban Runoff Program reported higher TSS ranging from 141 to 224 mg/L for stormwater in a "median urban site"

Table 3

Mean adsorption coefficient (K_d) values and standard deviations for bifenthrin, along with mean % of pyrethroid residues associated with the suspended solids (SS) phase, total suspended solids (TSS), and dissolved organic carbon (DOC) for bifenthrin in each sampling region.

County	n	Bifenthrin K_d ($\times 10^5$ L/kg)	% in SS	TSS (mg/L)	DOC (mg/L)
Sacramento/Yolo	36	2.4 ± 4.2	58.3	28.6	51.2
Alameda	27	1.9 ± 1.9	55.6	16.8	35.8
Madera	34	1.7 ± 1.8	37.9	35.4	30.5
Tulare	29	1.6 ± 1.7	48.3	31.8	51.6
Los Angeles	24	1.4 ± 2.6	48.9	54.5	59.3
Orange	36	3.9 ± 12.4	44.3	71.5	54.3
San Bernardino	19	1.6 ± 1.1	36.7	22.2	78.1
All regions	205	2.1 ± 5.6	47.7	38.6	49.8

(Morgan et al., 2005; Pitton et al., 2016; USEPA, 1983). In this study, the mean DOC level in catch basin water was 50 ± 42 mg/L and the median was 44 mg/L. This mean is similar to that from sediment pore water from San Diego Creek Watershed samples (42 mg/L), suggesting a strong contribution of sediment and organic debris to the elevated levels of DOM in the catch basin water (Budd et al., 2007). Generally, a higher TSS results in increased pyrethroid partitioning to the solid phase, and a higher DOC results in higher partitioning to dissolved organic matter in the aqueous phase (Liu et al., 2004). Suspended solids tend to settle to the bottom of a catch basin under low flow conditions, while DOM and the associated contaminants may travel over a longer distance and contaminate downstream surface water bodies. However, neither TSS nor DOC were found to be associated with pyrethroid detection or concentrations, indicating the influence of other factors on occurrence.

To better understand pyrethroid partitioning between the solid and aqueous phases, the partition coefficient K_d (L/kg) was calculated for bifenthrin residues. The average K_d for bifenthrin across all samples was 2.15×10^5 L/kg, with individual regions averaging from $1.42 \pm 2.6 \times 10^5$ L/kg in Madera County to $3.85 \pm 12.4 \times 10^5$ L/kg in Orange County (Table 3). These values are on a similar order of magnitude to those observed in a previous study (2.4×10^3 to 1.1×10^5 L/kg) where pyrethroids in runoff water from concrete surfaces were characterized following simulated precipitation events (Jiang and Gan, 2012). While pyrethroids are highly hydrophobic in general (log K_{ow} ranging from 4.5 to 7.0), previous studies using soils or sediments at equilibrium conditions have often found smaller K_d values, suggesting that suspended solids, likely due to their larger specific surface areas and high organic carbon content, have a stronger affinity for pyrethroid residues (Delgado-Moreno et al., 2010; Fojut and Young, 2011; Gan et al., 2005; Laskowski, 2002; Yang et al., 2006).

Though the high apparent K_d values would imply enrichment of pyrethroid residues on the suspended solids, a significant portion of the whole-water pyrethroids was still associated with the aqueous phase. An average of 47.7% of the pyrethroid residues was associated with suspended solids, leaving more than 50% in the aqueous phase (Table 3). Pyrethroid interactions with DOM may decrease distribution to the solid phase, in turn enhancing off-site transport. In a study of DOC from various media, the mean dissolved organic carbon-water partition coefficient K_{DOC} (L/kg) for bifenthrin ranged from 4.8×10^4 to 25.0×10^4 L/kg, indicating a strong affinity of pyrethroids for DOM (Delgado-Moreno et al., 2010). Findings from this and previous studies together suggest that, for strongly hydrophobic contaminants like pyrethroids, DOM facilitates short-term offsite transport, while solid particles, particularly after settling, may constitute a source for sustained emission in urban environments containing both artificial (e.g., USDS) and natural compartments (Jiang et al., 2016, 2010; Jiang and Gan, 2012; Richards et al., 2016).

Depending on the sedimentation rate in each catch basin, which may be influenced by factors like catch basin structure, particle size, water

flow rate, much of the suspended solids may settle at the bottom of catch basins and act as a reservoir for pyrethroid residues. Runoff may result in scouring, where sediment is resuspended, which has the potential to carry the contaminant-laden solids downstream and eventually into urban aquatic ecosystems. For flow rates less than $0.03 \text{ m}^3/\text{s}$, which is likely the case for the most of the dry season, a high proportion of the sediment may be captured by catch basins (Yang et al., 2018). A study using a physical catch basin model showed that, under its highest experimental flow rate of $0.01 \text{ m}^3/\text{s}$, the sediment was resuspended at a rate of 128 g/min when there was an overlaying water depth of 0.01 m (Avila et al., 2011). Under this scenario, assuming pyrethroid concentrations in the settled sediment were similar to those in the suspended solids, a median catch basin would have approximately 300 $\mu\text{g}/\text{min}$ of residues being re-suspended and available for offsite transport. However, increased flows from rain-induced runoff may pose a high risk of scouring. In California, this problem may be exacerbated by pollutant buildup over a long dry period, resulting in a more significant first flush when the rainy season begins (Lee et al., 2004). Climate models predict an increase in extreme precipitations for California; thus, instances of high flow and scouring may become more frequent, further highlighting the significance of catch basins as a substantial secondary source for contaminant redistribution in urban environments (Polade et al., 2017).

3.3. Potential effects on non-target aquatic organisms

The potential toxicity to freshwater invertebrates from whole water bifenthrin concentrations was estimated by calculating toxic units (TUs) based on a previously reported median 96-h EC_{50} (3.3 ng/L) and LC_{50} (7.7 ng/L) for *Hyalella azteca*, a freshwater amphipod commonly used for toxicity assessments (Table 4) (Weston and Jackson, 2009). Based on the EC_{50} , 89% of samples (all samples with concentrations above the reporting limit) showed $\text{TU} > 1$. Based on LC_{50} , 72% of samples contained bifenthrin at a level with $\text{TU} > 1$. Among all regions, the lowest average TU based on LC_{50} was 1.1, suggesting a threat to sensitive invertebrates from drainage water originating from the catch basins. It must be noted that the reporting limit for bifenthrin in the aqueous phase (2.0 ng/L) is somewhat close to this LC_{50} and may contribute to the low number of samples with $\text{TU} < 1$ for bifenthrin. Previous research has shown sediment from multiple sites in California impacted by urban runoff to be toxic to *H. azteca*, with bifenthrin being the primary driver of toxicity; though other pyrethroids, including permethrin, cyfluthrin, and cypermethrin, were also implicated (Amweg et al., 2006; Budd et al., 2020; Weston et al., 2005). While these TU estimates for catch basins are not representative of toxicity at stormwater outfalls or further downstream water bodies, they nevertheless highlight that sediments and water in the numerous catch basins, along with the USDS connecting these catch basins, may act as a significant source contributing to aquatic toxicity in urban watersheds.

In addition to toxicity to non-target organisms, inadvertent exposure to pyrethroids may select for pesticide resistance in populations of pests. Many pests that pose threats to public health and agriculture breed in

habitats that receive runoff, and resistance development in these species may hamper the efficacy of pest management practices. Resistant organisms also have the potential to bioaccumulate greater amounts of contaminants, increasing the risk of trophic transfer along the food chain (Johanif et al., 2021; Muggelberg et al., 2017). Though it is difficult to directly associate pesticide use with resistance, high agricultural use of pyrethroids has been linked to increased incidence of resistant pests, like mites and beetles, that can cause significant damages (Heimbach and Müller, 2013; Umina, 2007). For example, Hien et al. found resistant populations of the mosquito *Anopheles gambiae* and water contaminated with deltamethrin and lambda-cyhalothrin at sites of Burkina Faso in West Africa, suggesting selection pressure from environmental contamination as a result of cotton insecticide applications (Hien et al., 2017). Orondo et al. found increased phenotypic resistance in the mosquito *An. arabiensis* in irrigated areas of Kenya, where pyrethroids are the primary insecticides used for crop protection (Orondo et al., 2021). In California, the prevalence of a single mutation conferring pyrethroid resistance in *Culex pipiens* and *Cx. quinquefasciatus* was found to be nearly 1.5 times greater in populations from 2014 to 2016 as compared to those collected before 2012 (Yoshimizu et al., 2020).

Catch basins themselves are common breeding habitats for several mosquito species. USDSs, frequently containing standing water and food sources like algae and detritus, are a significant source of mosquitoes depending on the region, such as *Cx. quinquefasciatus* in southern California (Harbison et al., 2009; Kluh et al., 2006, 2001). Catch basins and other storm drain structures often contain stagnant water rich in organic matter, and are isolated from wind and sunlight, providing favorable conditions for mosquito eggs and larvae. Exposure to sublethal pyrethroid concentrations results in increased resistance in later generations of larvae, and larval resistance has been associated with adult resistance (Kawada et al., 2009; Shi et al., 2015). Therefore, the importance of urban catch basins in facilitating insecticide resistance in vector insects may constitute another impetus for better understanding the transport and fate of pyrethroids and other pesticides in USDSs and catch basins.

3.4. Mitigation of pyrethroid occurrence in catch basins

The ubiquitous presence of pyrethroid residues in urban catch basins, and the sheer number of catch basins in many highly urbanized areas, necessitate mitigation practices to minimize this overlooked source of contamination. Apart from directly reducing the use of pyrethroid insecticides, there are a number of practices that may be considered for reducing pyrethroid contamination in catch basins and storm drain systems. For example, smart irrigation systems would limit the generation of runoff from lawns and gardens. Many basin mouths that accept water from street gutters are completely open, meaning that the majority of sediments and small debris that can carry contaminants may enter freely (Azah et al., 2017; Jang et al., 2010). Covers and other amendments which are designed to prevent and remove large debris may also help reducing loading of contaminants that have a high affinity for solids, including pyrethroids (Alam et al., 2018a, 2018b; 2017;

Table 4

Toxic units (TUs) for *Hyalella azteca* from mean bifenthrin concentrations in whole water samples, grouped by county. SY = Sacramento/Yolo, AL = Alameda, MA = Madera, TU = Tulare, LA = Los Angeles, OR = Orange, SB = San Bernardino.

Month	TU endpoint	Region						
		SY	AL	MA	TU	LA	OR	SB
July	$\text{EC}_{50}^{\text{a,b}}$	36.5	19.2	16.6	19.8	15.3	17.8	12.9
	$\text{LC}_{50}^{\text{a}}$	15.6	8.2	7.1	8.5	6.6	7.6	5.5
August	EC_{50}	20.8	6.1	6.9	22.3	41.9	13.2	7.4
	LC_{50}	8.9	2.6	2.9	9.6	18.0	5.7	3.2
September	EC_{50}	7.4	12.0	15.2	5.8	110.5	2.7	5.6
	LC_{50}	3.2	5.2	6.5	2.5	47.3	1.1	2.4

^a Values for EC_{50} and LC_{50} derived from (Weston and Jackson, 2009).

^b Sublethal end point for EC_{50} defined as impaired swimming.

Morgan et al., 2005). Regular servicing of catch basins and other storm drain structures to remove the accumulated sediment and other debris, especially before the onset of the rainy season, is expected to greatly reduce the chemical loads from the first flush of rainfall-induced runoff. Physical catch basin dimensions vary widely and likely influence the fate of hydrophobic contaminants. Characteristics like height, sump depth, and outgoing channel diameter, may affect sediment capture and release into the USDS (Yang et al., 2018). For example, higher outlets, which may increase the maximum depth of water above the sediment, may reduce the amount of sediment that is resuspended during high flow (Avila et al., 2011). Storm drain system design that maximizes sediment capture would in turn limit the release of contaminants downstream.

The use of pesticides in urban areas is made by licensed operators as well as homeowners (Xie et al., 2021). While pesticide products have detailed instructions on the rate and method of application, there are often overuses with the intention to ensure complete pest eradications. In addition, pesticide misuses, including applications in non-infested areas like walkways and driveways, as well as spills from careless handling, may contribute greatly to contamination in surface runoff. Education on environmentally responsible pesticide use and handling should not target pesticide application professionals, but also the general public, to achieve meaningful reductions in offsite contamination of pesticides. The adoption of low-risk control methods, such as enclosed baits, as well as non-chemical methods, should be promoted to further minimize pesticide input in the environment.

4. Conclusions

The present study sought to determine the occurrence and patterns of the widely used pyrethroid insecticides in urban catch basins that receive runoff from irrigation and rainfall-induced runoff. From analysis of water from 79 catch basins across 8 counties in California during the dry season, nearly every sample contained one or more pyrethroids at a concentration above the reporting limit. Bifenthrin was a pyrethroid compound with the highest detection frequency as well as concentrations eliciting toxicity to sensitive aquatic invertebrates, although several other pyrethroids also contributed significantly to the chemical profile in catch basin water. Pyrethroid concentrations in catch basins generally decreased over time in the dry season, but high levels of residues remained in the aqueous and suspended solid phases at the end of summer, suggesting that residues in catch basins could serve as a significant secondary source for downstream contamination of urban surface streams and estuaries; the risk is expected to be the greatest during the initial phase of the rainy season, when rainfall-induced runoff may resuspend and mobilize pyrethroid residues retained in the catch basins. Pyrethroid pollution in urban regions adversely affect sensitive aquatic organisms and may also encourage resistance development in urban pests such as mosquitoes. To improve understanding of pesticide behaviors in urban catch basins and USDSs, further research should characterize runoff before and after it passes through a catch basin, the hydraulic retention of both water and solids in catch basins, and release of water and suspended solids from catch basins into the rest of the USDS. Efforts to design catch basins with reduced contaminant accumulation potential, and pest management practices to prevent the transport of pesticide residues from entering USDSs and catch basins, should be further explored.

Author Statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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